In the Specification

Kindly replace paragraphs [0001] through [0002] with the following:

Related Application

This is a §371 of International Application No. PCT/JP2005/004014, with an international filing date of March 2, 2005 (WO 2005/090069 A1, published September 29, 2005), which is based on Japanese Patent Application No. 2004-084836, filed March 23, 2004.

Technical Field

The present invention relates to an adhesion-enhanced polyimide film which is useful, as an electrical or electronic material, in the fields of electrical and electronic devices and semiconductors, which exhibits a sufficient practical level of peel strength for a laminated body obtained by lamination with a metal foil via an adhesive layer, and which has excellent dynamic characteristics, as well as to a process for its production and to a laminated body produced from it.

Background ~~Art~~

Polyimides, which have excellent properties including heat resistance, dimensional stability, dynamic characteristics, electrical properties, environmental resistance and flame retardance, as well as high flexibility, are widely used as flexible printed boards and tape automated bonding boards employed for mounting of semiconductor integrated circuits. In these fields, polyimide films are used as insulating supports for laminated bodies produced by lamination with metal foils, such as copper foils, via adhesives.

Kindly replace paragraphs [0010] through [0019] with the following:

~~Disclosure of the Invention~~

Nevertheless, there is much need of improvement because these polyimide films do not always provide sufficient peel strength to laminated bodies and, especially, to laminated bodies

prepared by lamination with metal foils such as copper foils via adhesives, while organic materials other than polyimides are problematic due to inadequate mechanical properties (tensile modulus) or thermal properties (linear expansion coefficient).

Summary

~~It is therefore an object of the present invention to~~We provide a polyimide film which provides a practical level of peel strength in laminated bodies obtained by lamination with metal layers, and which exhibits adequate mechanical properties (tensile modulus) or thermal properties (linear expansion coefficient) as a film, as well as a process for its production and laminated bodies produced from it.

~~The invention relates to~~We provide an adhesion-enhanced polyimide film which comprises a core layer composed of a polyimide (A) having high rigidity and a low linear expansion coefficient, at least one side of which has a thin layer formed by heating a coated layer comprising a heat-resistant surface treatment agent and a polyimide precursor which yields an amorphous polyimide (B).

~~The invention~~We further ~~relates to~~provide a process for production of an adhesion-enhanced polyimide film, wherein an organic solvent solution comprising a heat-resistant surface treatment agent and a polyimide precursor, which yields an amorphous polyimide (B) thin layer, is coated onto at least one side of a self-supporting film obtained from a polyimide precursor solution which yields a polyimide (A) core layer having high rigidity and a low linear expansion coefficient, to form a multilayer self-supporting film which is then heated and dried to complete imidation.

~~The invention~~We still further ~~relates to~~provide an adhesion-enhanced polyimide film which is obtained by the aforementioned production process.

~~The invention still further relates to~~We also provide a flexible metal layer laminated body comprising a metal layer laminated directly or via an adhesive onto the aforementioned adhesion-enhanced polyimide film.

~~According to the invention it~~It is possible to obtain a polyimide film having enhanced adhesion while retaining the excellent properties of the polyimide film.

~~According to the invention it~~It is also possible to easily obtain the aforementioned polyimide film, and to obtain laminated bodies with adequate peel strength.

~~Best Mode for Carrying Out the Invention~~Detailed Description

~~Preferred embodiments of the invention are those having~~Selected, representative aspects include the following ~~aspects.~~:

1) The aforementioned adhesion-enhanced polyimide film, wherein the polyimide (A) is obtained from 3,3',4,4'-biphenyltetracarboxylic dianhydride and p-phenylenediamine or p-phenylenediamine and 4,4'-diaminodiphenyl ether, from 3,3',4,4'-biphenyltetracarboxylic dianhydride and pyromellitic dianhydride and p-phenylenediamine or p-phenylenediamine and 4,4'-diaminodiphenyl ether, or from pyromellitic dianhydride and p-phenylenediamine and 4,4'-diaminodiphenyl ether.

2) The aforementioned adhesion-enhanced polyimide film, wherein the polyimide (A) is obtained using 3,3',4,4'-biphenyltetracarboxylic dianhydride and p-phenylenediamine as the main components (at 50 mole percent or greater to 100 mole percent of the total).

3) The aforementioned adhesion-enhanced polyimide film, wherein the polyimide (B) is obtained from at least one aromatic tetracarboxylic dianhydride selected from 2,3,3',4'-biphenyltetracarboxylic dianhydride, 2,2',3,3'-biphenyltetracarboxylic dianhydride, bis(3,4-

dicarboxyphenyl)ether dianhydride, bis(2,3-dicarboxyphenyl)ether dianhydride and naphthalenetetracarboxylic dianhydride, and an aromatic diamine.

4) The aforementioned adhesion-enhanced polyimide film, wherein the aromatic diamine is at least one member selected from p-phenylenediamine and 4,4'-diaminodiphenyl ether.

5) The aforementioned adhesion-enhanced polyimide film, wherein the heat-resistant surface treatment agent is an aminosilane compound, an epoxysilane compound or a titanate compound.

6) The aforementioned adhesion-enhanced polyimide film, wherein the polyimide (A) core layer has a thickness of 10-35 μm.

7) The aforementioned adhesion-enhanced polyimide film, wherein the polyimide (B) thin-layer has a thickness of 0.05-1 μm.

8) The aforementioned adhesion-enhanced polyimide film, wherein the polyimide film as a whole has a tensile modulus (MD) of between 6 GPa and 12 GPa and a linear expansion coefficient of 5×10^{-6} to 30×10^{-6} cm/cm/°C (at 50-200°C), preferably 10×10^{-6} to 30×10^{-6} cm/cm/°C (at 50-200°C).

It is an essential aspect of the process ~~of the invention~~ that an organic solvent solution comprising a heat-resistant surface treatment agent and a polyimide precursor, which yields a thin layer composed of an amorphous polyimide (B), is coated onto at least one side of a self-supporting film obtained from a polyimide precursor solution which yields a core layer composed of a polyimide (A) having high rigidity and a low linear expansion coefficient, to form a multilayer self-supporting film which is then heated and dried to complete imidation, and it is thereby possible to obtain an adhesion-enhanced polyimide film exhibiting a high practical level of peel strength for a laminated

body obtained by lamination with a metal foil via an adhesive layer, as well as adequate mechanical properties (tensile modulus) and thermal properties (linear expansion coefficient) for the film as a whole.

Kindly replace paragraph [0030] with the following:

The heat loss of the self-supporting film is the value determined by drying the film to be measured at 420°C for 20 minutes, measuring the weight before drying W1 and the weight after drying W2 and performing calculation by the following formula[[.]]:

Heat loss (wt%) = ((W1-W2)/W1} x 100.

Kindly replace paragraph [0033] with the following:

According to the process ~~of the invention~~, an organic solvent solution comprising a heat-resistant surface treatment agent and a polyimide precursor which yields an amorphous polyimide (B) thin layer is coated onto at least one side of a self-supporting film obtained from a polyimide precursor solution, which yields a polyimide (A) core layer, to form a multilayer self-supporting film.

Kindly replace paragraph [0035] with the following:

~~According to the invention, it~~It is essential to combine the heat-resistant surface treatment agent with a polyimide precursor which yields a thin layer of the amorphous polyimide (B). If either the heat-resistant surface treatment agent or a polyimide precursor which yields a thin layer of the amorphous polyimide (B) is used alone, the resulting polyimide film will not have adhesion enhanced to a practical level and, particularly, the adhesion will be too low for practical use if the thin-layer film has a thickness of less than 40 μm.

Kindly replace paragraph [0043] with the following:

~~According to the invention, the~~The sol solution obtained by the method described above may be coated onto at least one side (or both sides if necessary) of the self-supporting aromatic polyimide

precursor film by a publicly known coating method such as gravure coating, spin coating, silk screen coating, dip coating, spray coating, bar coating, knife coating, roll coating, blade coating, die coating or the like.

Kindly replace paragraph [0045] with the following:

The multilayer polyimide film ~~of the invention~~ is an adhesion-enhanced polyimide film which comprises a core layer composed of a polyimide (A) having high rigidity and a low linear expansion coefficient, at least one side of which has a thin layer formed by heating a coated layer comprising a heat-resistant surface treatment agent and a polyimide precursor which yields a highly heat-resistant amorphous polyimide (B), and preferably the polyimide (A) core layer has a thickness of 10-35 μm and the polyimide (B) thin layer has a thickness of 0.05-1 μm.

Kindly replace paragraph [0047] with the following:

The multilayer polyimide film ~~of the invention~~ is preferably used as a base film for a laminated flexible metal foil laminated body or a sputtered metal clad laminated body, or as a base film for a metal vapor deposited film, and especially as the base film of a flexible metal foil laminated body.

Kindly replace paragraph [0049] with the following:

The polyimide film ~~of the invention~~ may be used directly or, if necessary, after subjecting the thin-layer surface to corona discharge treatment, low-temperature plasma discharge treatment or ordinary-pressure plasma discharge treatment, for lamination with a metal foil via an adhesive to obtain a laminated body.

Kindly replace paragraph [0053] with the following:

A laminated body ~~according to the invention~~ may be easily produced in a continuous manner by, for example, coating the thin-layer surface of a polyimide film of the invention with a solution of

the aforementioned adhesive and drying the coated layer at a temperature of about 80-200°C for a period from 20 seconds to 100 minutes to form a thin film of the adhesive from which the solvent has been substantially removed to 1 wt% or less and preferably a solvent residue of no greater than 0.5 wt% (with a dry film or sheet thickness of about 1-20 μm), or by coating and drying a solution of the adhesive onto a resin film made of an aromatic polyester, polyolefin or the like to form a thin-film layer of the adhesive, subsequently combining it with the transferring thin-film layer side of the polyimide film of the invention to transfer the adhesive thin-film layer, forming a laminated body (sheet) with a metal foil such as a rolled copper foil, electrolytic copper foil, aluminum foil, stainless steel foil or the like, preferably with a rolled copper foil or electrolytic copper foil, and, in the case of a thermosetting adhesive, for example, performing lamination at a temperature of 80-200°C and especially 100-180°C under pressurization (0.2-50 kg/cm^2) followed by heating in a temperature range of 150-250°C for 1-24 hours for thermosetting, or in the case of a thermoplastic adhesive, for example, performing lamination at a temperature above the softening point of the adhesive and no higher than 350°C, under pressurization (0.2-100 kg/cm^2).

Kindly replace paragraph [0060] with the following:

~~The present invention~~Selected, representative aspects will now be explained in greater detail by examples and comparative examples.

Kindly delete paragraph [0097] as follows:

~~Industrial Applicability~~

~~The present invention is highly useful for industry as it provides a metal oxide thin film laminated polyimide film with excellent interlayer, cohesion and dynamic properties.~~